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Synthesis of Benzimidazole Derivatives: Microwave Approach review

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Abstract: Benzimidazole and its derivatives have very diverse biological activity. They have various biological activities like antimicrobial, antiviral, Antidiabetic, anticancer, activity. Different Benzimidazoles derivatives in good yield have been prepared by condensation of orthophenyldiamine with aromatic aldehyde in presence of various catalysts. In this we have reviewed large number of microwave assisted reaction involving synthesis of Benzimidazole derivatives..

Keywords: Benzimidazole, microwave, reaction, catalyst

I. INTRODUCTION

Heterocyclic compounds are compounds that contain one or more than one cyclic rings with one or more heteroatom/s like N, O, S or other in the ring. The forms of proteins like purine, histitidine, proline, and pyrimidine, bases in the genetic material like DNA and RNA are more important, which play vital role in life processes such as metabolism of all living cells. They also play important roles as enzyme, coenzyme and many natural products. Most biologically active molecules such as hormones, acids, enzymes, neurotransmitters may contain one, two or many heterocyclic rings. Out of these most common heterocyclic compound is Benzimidazole as an important nucleus.

There are a large number of pharmacologically active heterocyclic compounds as Benzimidazole, many of which are in regular clinical use. Very wide range of synthetic and naturally occurring heterocyclic compounds are having applications in medicines and also in pharmacy, pesticides, agrochemicals, polymers plastic ,drugs and dyes. There is a vast scope for the research leading to new heterocyclic molecules having very good biological activity.

The Benzimidazole derivatives were first realized as chemotherapeutic agents in 1950. Most common Benzimidazole containing compounds are 5, 6- dimethyl-1-D-ribofuranosyl Benzimidazole, part of the structure of Vitamin B-12 and other pharmaceutical drugs. The anthelmintic activity [1-2]are commonly known. Along with this, Benzimidazole derivatives with different pharmacological properties such as, anti-ulsaral[3-5], cardio tonic [6], antihypertensive against depression[7], antibacterial and antiviral against virus and bacteria [8], antitumor[9], antimutagent[10], antiallergenic[11] are already reported. It also exhibits analgesic, anti-inflammatory and antipyretic activity [12]. Moreover, it also shows hypoglycaemic [13],anticalmodulin [14] anti-aggregate [15] activities. Due to this synthesis of Benzimidazole derivatives.

II. IMPORTANCE OF BENZIMIDAZOLE RING SYSTEM:

A wide range of Benzimidazole and their derivatives find uses in pharmaceutical and veterinary drugs showing therapeutic activities. Some of the commercially important Benzimidazole derivatives are listed below.

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	Table 1.								
Sr. No.	Trade Name	Structure	Activity						
1	Cambendazole		Anthelmintic						
2	Albendazole		Anthelmintic						
3	Omeprazole		Anti-ulcer drugs						
4	Rabeprazole		Anti-ulcer drugs						
5	Esomeprazole		Anti-ulcer drugs						
6	Quinoline Benzimidazole analog		Anti- psychotic agents						
7	Imidazole derivative with Benzimidazole		Anti- psychotic agents						
8	Oxazolederivative with Benzimidazole	$ \begin{array}{c} & & \\ & & $	Antimicrobial activity						
9	Bibenzoimidazole derivatives		Antagonist						

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10	Benzimidazoleand coumarine derivative		Antiseptic activity
11	Amide derivative of Benzimidazole		Anticancer activity
12	Substituted Benzimidazole		Anticancer activity
13	Alkyl substituted Benzimidazole	N H H	Antiamoebic activity
14	Benzyl substituted carboxyl Benzimidazole		Antilukamic activity
15	Phenyl amine derivative of Benzimidazole	NH ₂ N N H	Antidiabetic activity
16	Amide derivative of Benzimidazole		Cyticidal activity
17	Oxfendazole		Roundworms and tapeworms
18	Ricobendazole	0 ² N O O	Anthelmintic
19	Triclabendazole		Anthelmintic

Besides the very high efficiency for the synthesis of Benzimidazoles, many of the methods required to be improved for the very high reaction temperature, very long reaction times, highly toxic solvent and high-cost catalyst, etc. [16] Therefore, there is need of development of simple mild, efficient and environmentally benign protocol for the synthesis of Benzimidazoles is still a hot topic for researchers. After the first reports of applications of microwaves in synthetic chemistry in 1986, now-a-days microwave-assisted, synthesis has become popular, particularly, last two decades due to the generally short reaction times, the high purity and yields of the resulting products with high purity. Up to now,

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several microwave-assisted methodologies for the synthesis of Benzimidazoles are reported in this review we have reported the different method for the synthesis of Benzimidazole derivatives using microwave reactions

1] Bougrin Kand etal

The microwave-assisted Benzimidazole synthesis by Bougrin They have described a Benzimidazole synthesis method using 1,2-diaminobenzene or 4-substituted-1,2-diaminobenzene and ethyl acetoacetate or ethyl benzoyl acetate on solid mineral supports or other support in dry media under microwave irradiation in domestic ovens leads to formation of the Benzimidazole derivative's.[17]



2] Emre Mentese and etal

Simple protocol was developed for the synthesis of Benzimidazoles. The reaction of iminoester hydrochlorides of phenylacetic with 4,5-dichloro-1,2-phenylenediamine or their derivative's under microwave irradiation leads to the Benzimidazole derivatives with good yields and in very short reaction times. [18]



3] D. Secci and etal

1, 2-diaryl-Benzimidazole and 2-aryl-1H-Benzimidazole derivatives were synthesized with simple differences using both microwave irradiation and conventional heating methods by carrier. Usually higher yields and time reactions reduction were obtained with the former method. Reaction requires very less time. [19]



4] Jyoti Pandey etal

Simple microwave assisted one-pot synthesis of Benzimidazoles from 1, 2 Phenylenediamine and aromatic aldehyde catalysed oxalic acid as a catalyst is described. Advantage technique, use the inexpensive and readily available catalyst, reaction time was decreased and the products were obtained in higher yields and having easy isolation. [20]



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5] B. Guruswamy et al

Ortho phenylene diamine is aromatic aldehyde acid was placed on H_2SO_4 -SiO₂ and transferred in to a microwave vial. The vial was sealed and placed in microwave. The reaction was run at 80 °C for 5 min. after completion of the reaction the product was obtained by the extraction and purification by column chromatography.[21]



6] Janardan Singh Yadav etal

The reaction of 2-acetyl Benzimidazoles with substituted aldehydes in methanol in presence of base afforded corresponding benzimidazolyl chalcones or unsaturated compounds which on treatment with ethylenediamine or phenyldiamine afforded the under MWI condition. Reaction occurs in short time and high yield.[22]



7] D.D. Rishiapathak etal

The 2-alkyl and 2-aryl substituted Benzimidazole derivatives is synthesized by reacting *o*-Phenylenediamine with various carboxylic acids using polyphosphoric acid as catalyst using by irradiation microwave methods. Reaction required very less time and good yield. [23]



8] Feng, F. etal

Synthesis of Benzimidazoles from 2-nitroaniline and ethanol over Cu-Pd/-Al₂O₃ catalysts are easily available starting materials, having high efficiency, and a simple procedure. The modification by Mg of the Cu-Pd/-Al₂O₃ catalyst could improve the catalytic activity significantly. Reaction carried in microwave at 100 w and gives better yield with good purity.[24]



9] Madhura Vijay Newrekar

The reaction of the From condensation of o-phenylene diamine and different substituted aromatic carboxylic acids and aromatic aldehydes, derivatives of 2-aryl Benzimidazole were synthesize using the microwave approach with good yield and purity.the reaction was catalysed by ammonium chloride and water [25]



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10] G. Navarrete and etal

2-(substituted phenyl)-1H-Benzimidazole derivatives with various 5-and 6-position substituents were synthesized via microwave irradiation using a short synthetic route and $Na_2S_2O_5$ as oxidant. This simple, fast, and efficient preparation of Benzimidazole derivatives has been developed using readily available and inexpensive reagents under solvent-free conditions. [26]



11] Limin Wang etal

An Efficient Procedure for the Synthesis of Benzimidazole Derivatives Using Yb(OTf)3 as Catalyst under Solvent-Free Conditions, in microwave Synthetic products obtained are purifi4d over silica gel.[27]



12] Abu Khan etal

Simple and efficient method for the convenient synthesis of 2-arylBenzimidazolehas been described on reaction with o-Phenylenediamine and various aromatic aldehydes using cobalt(II)chloride hexahydrate as a catalyst. The method is cost-effective, high-yielding, clean, and selective method for the synthesis.[28]



13] Na Zhao etal

An efficient and a quick microwave-assisted synthesis of Benzimidazoles and trisubstituted imidazole's was developed. Benzimidazoles were obtained as a result of the condensation of 1,2-phenylenediamine with carboxylic acids and acetoacetic ester without catalyst. Trisubstituted imidazoles were synthesized by condensation of benzil, aromatic aldehyde and ammonium acetate in the presence of glacial acetic acid, under microwave [29]



14] Hee-Jong Lim etal

Microwave-assisted condensation of resin-bound esters with 2-aminothiophenols to the corresponding benzothiazoles and BenzimidazoleThe condensation of ester with 2-aminothiophenol in 15% of methanesulfonic1, 2-dichlorobenzene system gave highest yield of 2-phenylbenzothiazoles Most esters including nicotinate ester were converted to the corresponding benzothiazoles and Benzimidazole [30]



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15] A. Saberi<u>et al</u>

Synthesis of Benzimidazole derivatives is outlined in. *o*-Phenylenediamine Carboxylic acid derivatives) and 50 mg of Alumina or Silica gel or Zeolite were mixed thoroughly in a mortar. The reaction mixture was then irradiated in a domestic microwave oven for 5-9 min at 160-560 W. The reaction gives good yield and purity.[31]



16] Jun Lu etal

In the presence of the PPA the mixture of acetic acid and o-phenylenediamine was stirred and irradiated in the MW oven three times at the 18% full output power (162 W). This reaction good yield and purity.[32]



17] Hossein Naeimi etal

Solution of o-phenylenediamine (and aldehyde in a minimum amount of acetonitrile was prepared. DDQ (0.14 g, 60 mol %) was added into an open Erlenmeyer flask. The mixture was irradiated in a microwave oven. The progress of the reaction was monitored by TLC. [33]



18] Bougrin K etal

Bougrin *et al*have described in dry media synthesis of 2-trifluoromethylBenzimidazoles through cyclocondensation of N-(carbotrifluoromethyl)-*ortho*-arylenediamines on montmorillonite K10 under domestic microwave oven with good yields[34]



19] Getvoldsen GS etal

Getvoldsen *et al.* method for the reactions in a microwave The Benzimidazole synthetic reaction from 1,2diaminobenzene and formic acid has chosen as a model reaction. They have reported that the new method would be an excellent analytical tool for monitoring the progress of a reaction.this gives good yield and purity.[35]



20] Boufatah N etal

Boufatah *et al*.have reported the preparation of some new biologically active Benzimidazole-4,7-dione derivatives in 7 steps through the microwave irradiation for the ring closing step to obtain Benzimidazole derivative [36]

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21] Getvoldsen Getal

Getvoldsen *et al* have reported 2-([4-F] fluorophenyl) Benzimidazole synthesis from the cyclocondensation reaction of 1,2-diaminobenzene with radiolabelled [4-F]f luorobenzoic acid in neat methanesulfonic and polyphosphoric acids under microwave [37]



22] Martinez-Palou R, etal

Martinez-Palou *et al.* (49) have described 2-long alkyl chain substituted Benzimidazole synthesis from the reaction of 1,2-diaminobenzene and stearic acid via mono and multimode microwave irradiation with high yields [38].



23] VanVilet DS etal

Vliet *et al.* have described a simple, rapid, one pot procedure for the generation of 2- substituted Benzimidazoles with high-yield directly from 2-nitroanilines using $SnCl_2$ as a reducing agent and carboxylic acid under microwave irradiation at 130 °C in 5 min [39]



24] Lin A-Y et al

Lin *et al.* have reported a microwave-assisted one-pot synthesis of several Benzimidazole derivatives from readily available starting compounds such as 1,2-diaminobenzene, 4,5- diaminopyrimidine, *cis*-1,2-diaminocicylohexane and several carboxylic acids including heteroaraomatic carboxylic acids .[40]





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25] Jing X et al

Jing *et al.* have described a simple and rapid synthesis of Benzimidazoles from 1,2-

diaminobenzene dihydrochloride and esters under microwave irradiation. This is also the first report for microwave assisted Benzimidazole synthesis from esters and 1,2-diaminobenzenes various Benzimidazole derivatives are synthesized by ethylene glycol as solvent [41]



26] Hosamani KM, etal

Similar to the Keri method, Hosamani and co-workers described a convenient protocol for the preparation of 5-nitro-2arylsubstituted phenoxymethyl-1*H*-Benzimidazole libraries in one pot-synthesis both under microwave irradiation and conventional heating methods using hydrochloride acid as catalyst [42]reaction is simple condensation reaction between orthophenyl diamine and acid functional group.



27] Hasaninejad A, etal

Hasaninejad *et al.* have reported the synthesis of some 2-substituted Benzimidazole derivatives from benzene-1,2diamine with mono and dicarboxylic acids under microwave irradiation using silphox [POCl3-n(SiO2)n] catalyst in highly yield and short reaction times [43]



28] Ben-Alloum A

Ben-Alloum *et al.* have described oxidative heterocyclization of aldehydes and ophenylenediamine with nitrobenzene or dimethylsulfoxide impregnated on silica gel irradiated with microwave in good yields and high purity [44].



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29] Zahran MA-H, etal

Zahran and co-workers described the synthesis of new heterocyclic compounds containing pyrazol-5-one coupled with Benzimidazole under dry media and they also explored antitumor activity of the newly synthesized heterocycles. Some of them were found to be more effective than thalidomide. [45]



30] Wen Xiaoan etal

Wen *et al.* have reported on eco-friendly, one-pot efficient propylphosphonic anhydride (T_3P) mediated synthesis of Benzimidazoles from various carboxylic acids and 1,2- diaminobenzene under microwave irradiation [46]



31] RS Joshi etal

Joshi *et al* (2010) employed microwave method for synthesising 2-ArylBenzimidazole. A mixture of substituted aldehyde, *o*-phenylene diamine and TBAF (5 moles %) was dissolved in minimum quantity of water with constant stirring and was irradiated under ultrasonic irradiation or microwave at ambient temperature for appropriate time. The progress of reaction was monitored by TLC. [47]



32] Niknam etal

Niknam et al (2007) reported microwave irradiation method as the best method for the synthesis, of 2-substituted Benzimidazoles and bis Benzimidazoles from the direct reaction of

phenylenediamine and dicarboxylic acid in the presence of alumina-methanesulfonic acid (AMA) as a catalyst with good to excellent [48]



33] R Dua etal

In a typical procedure, a mixture of 1,2-phenylenediamine(5mmol) carboxylic acid, alumina (0.5g) and methanesulfonic acid (12 mmol) was subjected to microwave irradiation (900 W, with a frequency 2450 MHz) at 20% power for times specified. [49]



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34] Singh et al

Singh *et al* (2010) synthesised 1, 3 dihydrobenzimidazol-2-thione and its derivatives in. 1,3 Dihydro-Benzimidazol-2-thione was formed by reacting *o*-phenylenediamine and thiourea in microwave at 40% intensity for 6 minutes 10 seconds till the colour changes to brown. Acetylchloride, chloroacetyl chloride and chlorosulfonic derivatives of 1,3, dihydro benzimidazol -2-thione (65) were synthesised from 1,3 Dihydro-Benzimidazol-2-thione . [50]



III. CONCLUSION

The Benzimidazole ring is an important pharmacophore in modern drug discovery component, also naturally occurring in vitamin B-12.Most of the Benzimidazole derivatives are synthesized by, heating, sonication or microwave energy. Most of these methods i) from the reaction of 1,2-diaminobenzenes with carboxylic acids or its derivatives, ii) condensation of 1,2-diaminobenzenes with aldehydes presence of an oxidative reagent. This method involves use of microwave reactions which require very short life time, good yield and purity. These methods are environmentally benign protocol for the synthesis of Benzimidazoles continues to attract researchers' attentions. This article aims to review the work reported, microwave-assisted synthesis of Benzimidazole derivatives.

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